LOW TEMPERATURE CARBONIZATION IN A FLUIDIZED BED

BY

HYMAN NATHAN STONE

B.S., University of New Hampshire, 1943
M.S., University of Illinois, 1947

AN ABSTRACT OF A THESIS

SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
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Introduction

The reaction kinetics of coal carbonization are inherently so complex that any study must be simplified. Much work has been done in fixed bed reactors, but it is difficult to determine the extent to which the reaction rates observed are controlled by the rate of heat transfer or the rate of gas diffusion through the bed. By using the fluid bed it is possible to minimize these effects.

The object of this work has been to obtain as much exploratory information as possible regarding the rates and mechanisms of carbonization in the low temperature range. Samples of non-caking bituminous coals were injected into a heated bed of silica, fluidized with pre-heated nitrogen. The course of the thermal reactions was traced by withdrawing, at specified time intervals, samples of solids and gases for analysis. The fluid bed of silica served primarily as a heat reservoir and the nitrogen was considered inert, so the reactions studied were simply those of thermal decomposition. Contact time, temperature, and the resulting products were investigated in two coals. Some work was also done on the effect of particle size.

The steady state residual volatile matter showed definite indications of being a linear function of carbonizing temperature. The data of other authors using the fluid bed technique were checked and found to follow this relationship.

Theoretical Considerations

Coal carbonization is believed to involve a series of thermal cracking reactions. Fuchs and Sandhoff (1) have calculated the free energy changes accompanying the formation of the split products found in the gases of carbonization. The results show that this mechanism of formation of the products evolved

at various temperatures is thermodynamically consistent. The rates of carbonization and the rate controlling mechanisms, however, have been partially obscured by the physical limitations of fixed bed studies. By using the fluid bed, coal samples can be injected into an inert preheated bed, fluidized with an inert preheated gas. The coal particles are immediately dispersed throughout the bed and rapidly and uniformly heated to the control temperature (2) (3). The volatile products are swept from the reaction zone by the fluidizing gas. In this manner, the limiting features of slow rates of heat transfer and product removal are avoided. Consequently, samples of solids withdrawn from the bed and samples of the noncondensible gases give a more intimate picture of primary carbonization. The composite picture observed in this manner is still complex, but it is possible to evaluate more closely the relative importance of various proposed controlling mechanisms. Fuchs and Sandhoff have proposed a unimolecular cracking reaction concept and have calculated activation energies based on it. Berkowitz (4) has advanced a theory that the physical structure and pore diameter determine the carbonizing characteristics. Sinnatt (5) and others (6) have done considerable work that indicates that the various banded ingredients react differently to heat.

Experimental Apparatus and Procedure

The reactor was a four foot stainless steel tube, 4.5 inch inside diameter. The bed was supported on a stainless steel porous plate of a 65 micron mean pore diameter. The coal was injected into the bed of silica by means of a high speed screw conveyor. The temperature was controlled with a Wheelco Xactline controller operated by a thermocouple inserted into the center of the bed. The samples of solids were analyzed by a modified A.S.T.M. proximate analysis and the gases were analyzed in a standard Burrell unit.

Results and Conclusions

The data obtained indicate that the rate of carbonization

is the result of a combination of several reactions that produce volatile products. This is also supported by microscopic examination which indicated that the components vitrain and clarain react differently to pyrolysis. In the particle size range studied, the particle size and porosity seem to have little effect upon the rate of devolatilization, except in the first 15 seconds. Measurements taken in the first 15 seconds are uncertain, since the coal charging time was 17 seconds.

The results show that the steady state volatile matter can be expressed as follows:

$$V = mt + b \qquad (1)$$

where V is the per cent volatile matter on an ash and moisture free basis and t is the carbonizing temperature in °F. The slope m appears to be independent of the coal used while the intercept b is a function of the coal and its previous oxidation history. The data of Singh and Kane (7) were found to fit this relationship also.

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VITA

The author was born on May 8, 1920, in Claremont, New Hampshire. He attended public schools and was graduated as valedictorian from Stevens High School in that community in 1937. He spent two years working and entered the University of New Hampshire in the fall of 1939. He was graduated with the Degree of Bachelor of Science in Chemical Engineering in May, 1943. The next three months were spent in the employ of the United States Bureau of Mines at College Park, Maryland, as a junior Chemical Engineer. He entered the United States Navy at that time and received training in electronic engineering at Princeton University and Massachusetts Institute of Technology. He then served as an electronics maintenance officer on several Naval Air Bases in the Pacific until his separation in May, 1946. At that time he entered the Graduate College at the University of Illinois and received the Degree of Master of Science in Chemical Engineering in October, 1947. He was the recipient of a Cogswell Scholarship at the University of New Hampshire. He worked as a teaching assistant and received a Pittsburgh Consolidation Coal Company Research Fellowship at the University of Illinois.